

Time evolution of an entangled initial state in coupled quantum dots with Coulomb correlations

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We analyzed the dynamics of the initial singlet electronic state in the two interacting single-level quantum dots (QDs) with Coulomb correlations, weakly tunnel coupled to an electronic reservoir. We obtained correlation functions of all orders for the electrons in the QDs by decoupling high-order correlations between localized and band electrons in the reservoir. We proved that for arbitrary mixed state the concurrence and entanglement can be determined from the average value of particular combinations of electron's pair correlation functions. Analysis of the pair correlation functions time evolution allows to follow the changes of concurrence and entanglement during the relaxation processes. We investigated the dependence of concurrence on the value of Coulomb interaction and the energy levels spacing and found it's non-monotonic behavior in the non-resonant case. We also demonstrated that the behavior of pair correlation functions for two-electron entangled state in coupled QDs points to the fulfillment of the Hund's rule for the strong Coulomb interaction. We revealed the appearance of dynamical inverse occupation of the QDs energy levels during the relaxation processes. Our results open up further perspectives in solid state quantum information based on the controllable dynamics of the entangled electronic states.

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I. INTRODUCTION

Now a days, low-dimensional semiconductor structures with a small number of electrons attract much attention. The main reason for that interest is the progress in technological procedure that allows to fabricate nanostructures with high precision [1],[2]. In recent years experimental technique gives possibility to create vertically aligned strongly interacting QDs with only one of them coupled to the continuous spectrum states [3],[4]. This side-coupled geometry gives an opportunity to fabricate many-particle states with various charge and spin configurations in the small size structures [5],[6],[7],[8],[9]. Considerable progress was achieved in fabrication of lateral QDs, which are extremely tunable by means of individual electrical gates [10],[11]. This advantage reveals in the possibility of single electron localization in the system of several coupled QDs [12] and charge states manipulations in the artificial molecules [13]. There are a lot of possible applications of this field in nanoelectronics [14], including quantum information processing [15]. Double QDs play an important role in the problem of quantum information processing [16],[17]. Most

of the proposed schemes for quantum computation deal with the spin control because of the long decoherence times [18]. But now due to the development of light sources the control on electric charge in low-dimensional systems is produced both by gate voltages [19],[20] and laser pulses [21],[22]. Creating, controlling and detecting entangled states in ultra small condensed matter systems is one of the most important problems for future quantum computation applications [23] and for the development of new electronic devices based on semiconductor nanostructures. It is important to be able to prepare interacting few-level systems with different initial states [24],[25],[26],[27],[28] - from simple product states to complex entanglements. Various ideas for entangling of spatially separated electrons were proposed, such as, by splitting Cooper pairs [29] or by spin manipulation in QDs [30],[31]. In double QDs an entangled state can be obtained by putting the electrons into a singlet ground state [16],[32],[33]. Electron transport in coupled QDs is governed by Coulomb interaction between localized electrons, by the ratios between tunneling transfer amplitudes and the quantum dots coupling and of course by the initial conditions [34],[35]. To integrate quantum dots in a small quantum circuits it is necessary to analyze the influence of non-equilibrium charge distribution, relaxation processes and non-stationary effects on the electron transport through the system. So the problem of charge relaxation due to the tunneling processes between QDs coupled to an electronic reservoir in the presence

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of Coulomb interaction is really vital. Consequently, the detailed analysis of time evolution of initial singlet entangled two-electron state in the system of interacting QDs with Coulomb correlations is an important problem, which may have further implications for quantum information in nanoscale devices.

In this paper we consider charge relaxation in the double QD due to the coupling to an electronic reservoir. Tunneling from the first QD to the continuum is possible only through the second dot. We obtained the closed system of equations for time evolution of the localized electrons filling numbers and pair correlation functions which exactly takes into account all order correlation for localized electrons. We decoupled the high order correlation functions between conduction electrons in the reservoir and electrons localized in the QDs. In such an approximation the electrons distribution in the reservoir is not influenced by changing of an electronic states in the coupled QDs. For QDs weakly coupled to the reservoir the proposed decoupling scheme is a good approximation. We considered system relaxation from initial singlet entangled two-electron state and took into account Coulomb correlations within both QDs. Such state can be prepared experimentally as a ground two-particle state for vertically aligned strongly coupled QDs if interaction with substrate is extremely weak in comparison with interdot coupling and interaction with the other lead (reservoir). The interaction with reservoir (for example STM tip) is switched on at the initial time moment. We proved that for a mixed state the concurrence and entanglement can be expressed through the average value of particular combinations of electron's pair correlation functions. We performed the analysis of the pair correlation functions time evolution, which allowed us to follow the changes of concurrence and entanglement during the relaxation process. We found some peculiarities in electrons filling numbers dynamics arising due to the Coulomb correlation effects. We demonstrated the appearance of dynamical inverse occupation in the proposed system and revealed the fulfilment of Hund's rule [36],[37],[38].

II. MODEL

We consider a system of coupled QDs with the energy levels ε_1 and ε_2 connected to an electronic reservoir (see Fig.1). At the initial time moment the interaction between the QD with energy level ε_2 (second QD) and electronic reservoir (ε_p) is switched on. In the absence of interaction with the reservoir two-electronic states in the coupled QDs are entangled in the presence of Coulomb correlations. We model the system by the Hamiltonian \hat{H} :

$$\hat{H} = \hat{H}_D + \hat{H}_{tun} + \hat{H}_{res}. \quad (1)$$

The Hamiltonian \hat{H}_D of interacting QDs

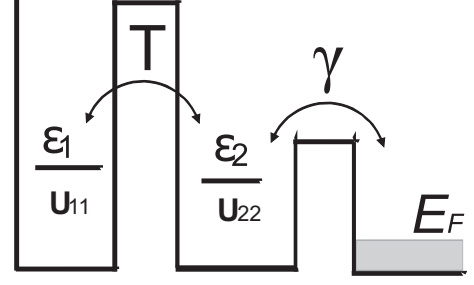


FIG. 1: Scheme of the proposed model. The system of interacting QDs is coupled to an electronic reservoir by means of tunneling rate $\gamma = \pi\nu_0 t^2$.

$$\begin{aligned} \hat{H}_D = & \sum_{l=1,2\sigma} \varepsilon_l c_{l\sigma}^\dagger c_{l\sigma} + \sum_{l=1,2\sigma} U_l n_{ll\sigma} n_{ll-\sigma} + \\ & + \sum_{\sigma} T (c_{1\sigma}^\dagger c_{2\sigma} + c_{2\sigma}^\dagger c_{1\sigma}), \end{aligned} \quad (2)$$

contains the spin-degenerate levels ε_l (indexes $l = 1$ and $l = 2$ correspond to the first and to the second QD) and the on-site Coulomb energy U_l for double occupation of the dots. The creation/annihilation of an electron with spin $\sigma = \pm 1/2$ within the dot is denoted by $c_{l\sigma}^\dagger/c_{l\sigma}$ and $n_{ll\sigma}$ is the corresponding filling number operator. The coupling between the dots is described by the tunneling transfer amplitude T which is considered to be independent of momentum and spin.

The reservoir is modeled by the Hamiltonian:

$$\hat{H}_{res} = \sum_{p\sigma} \varepsilon_p c_{p\sigma}^\dagger c_{p\sigma}, \quad (3)$$

in which $c_{p\sigma}^\dagger/c_{p\sigma}$ creates/annihilates an electron with spin σ and momentum p in the lead. The coupling between the second dot and the reservoir is described by the Hamiltonian:

$$\hat{H}_{tun} = \sum_{p\sigma} t (c_{p\sigma}^\dagger c_{2\sigma} + c_{p\sigma} c_{2\sigma}), \quad (4)$$

where t is the tunneling amplitude, which is considered to be independent on momentum and spin. Taking into account a constant density of states in the reservoir ν_0 , the tunnel coupling rate γ is defined as $\gamma = \pi\nu_0 t^2$. Interaction between the second QD and the reservoir is switched on at the initial time moment.

In the absence of coupling with the reservoir correlated two particle pure states for electrons with opposite spins are described by the wave function

$$|\psi\rangle = \alpha |\uparrow\downarrow\rangle|0\rangle + \beta |\downarrow\rangle|\uparrow\rangle + \gamma |\uparrow\rangle|\downarrow\rangle + \delta|0\rangle|\uparrow\downarrow\rangle. \quad (5)$$

For non-zero value of Coulomb interaction in the system this state is entangled. Such state with the lowest energy can be experimentally prepared in strongly

coupled vertically aligned QDs in the case when interaction with substrate is extremely weak in comparison with interdot coupling and interaction with the other lead (reservoir, for example STM tip).

We set $\hbar = 1$ and derive the kinetic equations for bilinear combinations of Heisenberg operators $c_{l\sigma}^+ / c_{l\sigma}$

$$\begin{aligned} c_{1\sigma}^+ c_{1\sigma} &= \hat{n}_{11}^\sigma(t); & c_{2\sigma}^+ c_{2\sigma} &= \hat{n}_{22}^\sigma(t); \\ c_{1\sigma}^+ c_{2\sigma} &= \hat{n}_{12}^\sigma(t); & c_{2\sigma}^+ c_{1\sigma} &= \hat{n}_{21}^\sigma(t). \end{aligned} \quad (6)$$

Kinetic equations describe time evolution of the electron filling numbers in each QD

$$\begin{aligned} i\frac{\partial}{\partial t} \hat{n}_{11}^\sigma &= -T(\hat{n}_{21}^\sigma - \hat{n}_{12}^\sigma), \\ i\frac{\partial}{\partial t} \hat{n}_{22}^\sigma &= T(\hat{n}_{21}^\sigma - \hat{n}_{12}^\sigma) - 2i\gamma \hat{n}_{22}^\sigma, \\ i\frac{\partial}{\partial t} \hat{n}_{21}^\sigma &= T(\hat{n}_{22}^\sigma - \hat{n}_{11}^\sigma) + [\xi + U_1 \hat{n}_{11}^{-\sigma}] \hat{n}_{21}^\sigma - \\ &\quad - U_2 \hat{n}_{21}^\sigma \hat{n}_{22}^{-\sigma} - i\gamma \hat{n}_{21}^\sigma, \\ i\frac{\partial}{\partial t} \hat{n}_{12}^\sigma &= -T(\hat{n}_{22}^\sigma - \hat{n}_{11}^\sigma) - [\xi + U_1 \hat{n}_{11}^{-\sigma}] \hat{n}_{12}^\sigma + \\ &\quad + U_2 \hat{n}_{12}^\sigma \hat{n}_{22}^{-\sigma} - i\gamma \hat{n}_{12}^\sigma, \end{aligned} \quad (7)$$

where $\xi = \varepsilon_1 - \varepsilon_2$ is the detuning between the energy levels in the QDs. The system of Eqs. (7) contains expressions for the pair correlators $\hat{n}_1^{-\sigma} \hat{n}_{21}^\sigma$ and $\hat{n}_1^{-\sigma} \hat{n}_{12}^\sigma$, which also determine relaxation and consequently have to be calculated. We'll neglect high order correlation functions between localized and reservoir electrons and fulfill averaging over electron states in the reservoir.

Let us introduce the following designation for the pair correlations operators $\hat{K}_{lr'l'r'}^{\sigma\sigma'}$ and their averaged values $K_{lr'l'r'}^{\sigma\sigma'} = \langle c_{l\sigma}^+ c_{r\sigma} c_{l'\sigma'}^+ c_{r'\sigma'} \rangle$. We'll consider only the paramagnetic case $\langle \hat{n}_l^\sigma \rangle = \langle \hat{n}_l^{-\sigma} \rangle$. The system of equations for pair correlators can be written in the compact matrix form (symbol $[\]$ means commutation and symbol $\{ \}$ - anti-commutation)

$$i\frac{\partial}{\partial t} \hat{\mathbf{K}} = [\hat{\mathbf{K}}, \hat{H}'] + \{\hat{\mathbf{K}}, \hat{\Gamma}\} + \hat{\Upsilon}, \quad (8)$$

where $\hat{\mathbf{K}}$ is the pair correlators matrix

$$\hat{\mathbf{K}} = \begin{pmatrix} K_{2211}^{\sigma-\sigma} & K_{1211}^{\sigma-\sigma} & K_{2221}^{\sigma-\sigma} & K_{1221}^{\sigma-\sigma} \\ K_{2111}^{\sigma-\sigma} & K_{1111}^{\sigma-\sigma} & K_{2121}^{\sigma-\sigma} & K_{1121}^{\sigma-\sigma} \\ K_{2212}^{\sigma-\sigma} & K_{1212}^{\sigma-\sigma} & K_{2222}^{\sigma-\sigma} & K_{1222}^{\sigma-\sigma} \\ K_{2112}^{\sigma-\sigma} & K_{1112}^{\sigma-\sigma} & K_{2122}^{\sigma-\sigma} & K_{1122}^{\sigma-\sigma} \end{pmatrix}, \quad (9)$$

matrix \hat{H}' has the following form

$$\hat{H}' = \begin{pmatrix} 0 & T & T & 0 \\ T & \xi + U_1 & 0 & T \\ T & 0 & -\xi + U_2 & T \\ 0 & T & T & 0 \end{pmatrix} \quad (10)$$

and the relaxation matrix $\hat{\Gamma}$ is denoted as

$$\hat{\Gamma} = \begin{pmatrix} -i\gamma & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & -2i\gamma & 0 \\ 0 & 0 & 0 & -i\gamma \end{pmatrix}. \quad (11)$$

It is clearly evident that Eqs. (8) contain expressions for the high-order correlators $K_{121122}^{\sigma-\sigma-\sigma}$ and $K_{211122}^{\sigma-\sigma-\sigma}$. Their contribution can be written in the matrix form $\hat{\Upsilon}$

$$\begin{pmatrix} 0 & U_2 K_{121122}^{\sigma-\sigma-\sigma} & U_1 K_{211122}^{\sigma-\sigma-\sigma} & 0 \\ -U_2 K_{211122}^{\sigma-\sigma-\sigma} & 0 & 0 & -U_2 K_{211122}^{\sigma-\sigma-\sigma} \\ -U_1 K_{121122}^{\sigma-\sigma-\sigma} & 0 & 0 & -U_1 K_{121122}^{\sigma-\sigma-\sigma} \\ 0 & U_2 K_{121122}^{\sigma-\sigma-\sigma} & U_1 K_{211122}^{\sigma-\sigma-\sigma} & 0 \end{pmatrix} \quad (12)$$

The system of equations (7),(8) for the two electronic pure entangled state $|\psi\rangle$ time evolution in the coupled QDs connected with the reservoir can be solved with the following initial conditions: $n_{11}^\sigma(0) = \alpha^2 + \beta^2$; $n_{12}^\sigma(0) = n_{21}^\sigma(0) = \alpha\gamma + \beta\delta$; $n_{22}^\sigma(0) = \delta^2 + \gamma^2$; $K_{1111}^{\sigma-\sigma} = \alpha^2$; $K_{2222}^{\sigma-\sigma} = \delta^2$; $K_{1122}^{\sigma-\sigma} = \beta^2$; $K_{2211}^{\sigma-\sigma} = \gamma^2$; $K_{1221}^{\sigma-\sigma} = K_{2112}^{\sigma-\sigma} = \beta\gamma$; $K_{2121}^{\sigma-\sigma} = K_{1212}^{\sigma-\sigma} = \alpha\delta$; $K_{1211}^{\sigma-\sigma} = K_{2111}^{\sigma-\sigma} = \gamma\alpha$; $K_{1112}^{\sigma-\sigma} = K_{1121}^{\sigma-\sigma} = \alpha\beta$; $K_{1222}^{\sigma-\sigma} = K_{2122}^{\sigma-\sigma} = \beta\delta$; $K_{2221}^{\sigma-\sigma} = K_{2212}^{\sigma-\sigma} = \gamma\delta$. The high-order correlators $K_{121122}^{\sigma-\sigma-\sigma}$ and $K_{211122}^{\sigma-\sigma-\sigma}$ are exactly equal to zero as they are the solution of the linear homogeneous system of equations with zero initial conditions.

Let us discuss the changing of entanglement properties of initial state during the system time evolution. A standard measure of the entanglement is the concurrence C [39],[40],[41]. For each pure state the entanglement E is defined as the entropy of either of the two subsystems A and B [42]:

$$E(\psi) = -\text{Tr}(\rho_A \log_2 \rho_A) = -\text{Tr}(\rho_B \log_2 \rho_B). \quad (13)$$

Here ρ_A is the partial trace of $|\psi\rangle\langle\psi|$ over subsystem B , and ρ_B has the similar meaning. The entanglement of the mixed state ρ is then defined as the average entanglement of the pure states of the decomposition, minimized over all decompositions of ρ :

$$E(\rho) = \min_i \sum_i p_i E(\psi_i). \quad (14)$$

To determine the concurrence for the system of two single-level QDs with two electrons with opposite spins one has to distinguish two different situations.

A. Subsystems A and B are QDs (I)

We now consider in detail the situation when subsystems A and B are QDs: the first QD - dot with energy level ε_1 and the the second QD - dot with energy level

ε_2 , directly coupled to the reservoir (subsystem C) correspondingly. Interaction between the second QD and the reservoir is switched on at the initial time moment $t = 0$. For the each dot four electronic states are possible: $|0\rangle_l$, $|\uparrow\rangle_l$, $|\downarrow\rangle_l$ and $|\uparrow\downarrow\rangle_l$, where $l = 1, 2$. We are going to analyze entanglement between the electronic states in subsystems A and B (the first and the second QD). The concurrence for pure state $|\psi\rangle$ is determined as $C_I = |\langle\psi|\tilde{\psi}\rangle|$, where $|\tilde{\psi}\rangle$ is the "spin flipped" state $|\psi\rangle$. For mixed state concurrence is $C_I = \max\{0, \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4\}$, where $\{\lambda_m\}$ - square roots of eigenvalues of matrix $\tilde{\rho}\rho$ ($\tilde{\rho}$ is the "spin flipped" matrix ρ) arranged in the decreasing order. For initial two-electron entangled pure state $|\psi\rangle$ [see Ex.(5)] with opposite spins one can define C_I

$$C_I = |\alpha^2 + \delta^2 + 2\beta\gamma|. \quad (15)$$

We'll demonstrate that for arbitrary mixed state concurrence C_I can be determined through the mean value of pair correlators $K_{lr'l'r'}^{\sigma-\sigma}$ particular combination

$$C_I = \langle K_{1111}^{\sigma-\sigma} + K_{1221}^{\sigma-\sigma} + K_{2112}^{\sigma-\sigma} + K_{2222}^{\sigma-\sigma} \rangle. \quad (16)$$

Let us introduce operator \hat{K}' , which can be expressed in terms of pair correlations operators:

$$\hat{K}' = \hat{K}_{1111}^{\sigma-\sigma} + \hat{K}_{1221}^{\sigma-\sigma} + \hat{K}_{2112}^{\sigma-\sigma} + \hat{K}_{2222}^{\sigma-\sigma}. \quad (17)$$

Acting by operator \hat{K}' on the wave function $|\psi\rangle$ [see Ex.(5)] one obtain "spin flipped" wave function $|\tilde{\psi}\rangle$

$$\hat{K}'|\psi\rangle = |\tilde{\psi}\rangle. \quad (18)$$

For any wave function $|\psi\rangle$:

$$\langle\psi|\hat{K}'|\psi\rangle = \langle\psi|\tilde{\psi}\rangle = C_I. \quad (19)$$

One can also find wave functions $\psi_i (i = 1, 2, 3, 4)$ for two-electron states with opposite spins, which are the eigenstates of the Hamiltonian \hat{H}_D

$$|\psi_i\rangle = \alpha_i |\uparrow\downarrow\rangle|0\rangle + \beta_i |\downarrow\rangle|\uparrow\rangle + \gamma_i |\uparrow\rangle|\downarrow\rangle + \delta_i |0\rangle|\uparrow\downarrow\rangle. \quad (20)$$

The corresponding eigenvalues E_i can be determined from equation

$$\det(\hat{H} - E\hat{I}) = 0, \quad (21)$$

where \hat{I} is the unity matrix and

$$\hat{H} = \begin{pmatrix} 2\varepsilon_1 + U_1 & T & -T & 0 \\ T & \varepsilon_1 + \varepsilon_2 & 0 & -T \\ -T & 0 & \varepsilon_1 + \varepsilon_2 & T \\ 0 & -T & T & 2\varepsilon_2 + U_2 \end{pmatrix}. \quad (22)$$

In the case of resonant tunneling between the similar QDs ($\varepsilon_1 = \varepsilon_2 = \varepsilon_0$; $U_1 = U_2 = U$) the coefficients α_i , β_i , γ_i and δ_i for the ground singlet state can be obtained analytically:

$$\begin{aligned} \alpha &= \delta = \frac{\sqrt{2}T}{\sqrt{4T^2 + b^2}}, \\ \beta &= \gamma = \frac{b}{\sqrt{2}\sqrt{4T^2 + b^2}}, \end{aligned} \quad (23)$$

where

$$b = \frac{U}{2} + \sqrt{\frac{U^2}{4} + 4T^2}. \quad (24)$$

The energy of the ground state has the value $E_G = \varepsilon_0 + \frac{U}{2} - \sqrt{\frac{U^2}{4} + 4T^2}$.

If $\{|\psi_i\rangle\}$ are the two-particle eigenfunctions for electrons with opposite spins of the Hamiltonian \hat{H} , two particle density matrix can be expressed as $\rho = \sum_i |\psi_i\rangle\langle\psi_i|p_i$. The following relations take place: $\langle\psi_j|\hat{K}'|\tilde{\psi}_i\rangle = \delta_{ij}$ and $\langle\psi_i|\hat{K}'^2|\psi_i\rangle = \delta_{ii'}$ = $\sum_j \langle\psi_i|\hat{K}'|\tilde{\psi}_j\rangle\langle\tilde{\psi}_j|\hat{K}'|\psi_i\rangle$.

Let us prove that

$$\langle\psi_j|\tilde{\rho}\rho|\psi_i\rangle = \langle\psi_j|\hat{K}'\rho\hat{K}'\rho|\psi_i\rangle. \quad (25)$$

Really:

$$\begin{aligned} \langle\psi_j|(\hat{K}'\rho)^2|\psi_i\rangle &= \sum_{i_1} \langle\psi_j|\hat{K}'|\psi_{i_1}\rangle\langle\psi_{i_1}|\hat{K}'|\psi_i\rangle p_i p_{i_1} = \\ &= \sum_{i_1} \langle\psi_j|\tilde{\psi}_{i_1}\rangle\langle\psi_{i_1}|\tilde{\psi}_i\rangle p_i p_{i_1} \end{aligned} \quad (26)$$

and

$$\langle\psi_j|\tilde{\rho}\rho|\psi_i\rangle = p_i \langle\psi_j|\tilde{\rho}|\psi_i\rangle = \sum_{i_1} p_i p_{i_1} \langle\psi_j|\tilde{\psi}_{i_1}\rangle\langle\tilde{\psi}_{i_1}|\psi_i\rangle \quad (27)$$

Taking into account expression (20) and comparing expressions (26) and (27), one can find that statement (25) is valid. If $\tilde{\lambda}_p$ are the eigenvalues of matrix $\|\tilde{\rho}\rho\|_{ji}$ and λ_m are the eigenvalues of matrix $\|\hat{K}'\rho\|_{ji'}$, then $\lambda_m^2 = \tilde{\lambda}_p$ and $\lambda_m = \pm\sqrt{\tilde{\lambda}_p}$. So,

$$Tr(\hat{K}'\rho) = \sum_m \lambda_m. \quad (28)$$

The concurrence C_I [40] is expressed through $\tilde{\lambda}_p$, arranged in decreasing order, as $C_I = \max\{0, \sqrt{\tilde{\lambda}_1} - \sqrt{\tilde{\lambda}_2} - \sqrt{\tilde{\lambda}_3} - \sqrt{\tilde{\lambda}_4}\}$. Finally

$$C_I = \max\{0, \text{Tr}(\hat{K}' \rho)\} = \max\{0, \langle \hat{K}' \rangle\}. \quad (29)$$

We would like to point out that for a pure state $|\psi\rangle$ [see Ex.(5)] the entanglement of subsystem A (first QD) with the surrounding subsystems B (second QD) and C (reservoir) can be expressed as:

$$E(\rho_A) = \text{Tr}_{BC} \rho_{ABC} = -\alpha^2 \log_2 \alpha^2 - \beta^2 \log_2 \beta^2 - \gamma^2 \log_2 \gamma^2 - \delta^2 \log_2 \delta^2 \quad (30)$$

In the absence of Coulomb interaction ($U = 0$) for symmetric QDs in the singlet two-electron state one can find that $\alpha = \delta = \beta = \gamma = 1/2$. In this case the concurrence C_I [see Ex.(15)] is equal to zero, but entanglement $E(\rho_A) = 2$. This means that subsystems A and B (first and second QDs) are disentangled, but electrons in the first QD are maximally entangled with the reservoir (subsystem C).

B. Subsystems A and B are opposite spin systems (II)

Let us now consider the other situation when the subsystem A corresponds to the electrons with spin projections $S_Z = +\sigma$ and subsystem B - to electrons with spin projections $S_Z = -\sigma$. Each particle with particular spin projection can be found in the first or in the second QD. If particle is found in the first QDs we attribute +1 to this state and if it is situated in the second QD we attribute -1. This case is similar to the problem of two interacting "frozen spins" at neighboring sites. For example, the states $|\uparrow\rangle|\uparrow\rangle$ and $|\downarrow\rangle|\downarrow\rangle$ in the "frozen spin" problem corresponds to the states $|\uparrow\downarrow\rangle|0\rangle$ and $|0\rangle|\downarrow\uparrow\rangle$ in the considered situation. The "spin flip" in the "frozen spin" problem corresponds to permutation of QDs. The pure state $|\psi\rangle$ [see Ex.(5)] is then transformed to the state $|\tilde{\psi}\rangle$

$$|\tilde{\psi}\rangle = \alpha|0\rangle|\downarrow\uparrow\rangle - \beta|\downarrow\rangle|\uparrow\rangle - \gamma|\uparrow\rangle|\downarrow\rangle + \delta|\uparrow\downarrow\rangle|0\rangle. \quad (31)$$

The concurrence C_{II} in the pure state can be determined as usual $C_{II} = |\langle \psi | \tilde{\psi} \rangle|$. For the state $|\psi\rangle$ [see Ex.(5)] the concurrence $C_{II} = |2\alpha\delta - 2\beta\gamma|$.

Let us introduce operator \hat{K}'' , which can be expressed in terms of pair correlations operators

$$\hat{K}'' = \hat{K}_{1212}^{\sigma-\sigma} + \hat{K}_{2121}^{\sigma-\sigma} - \hat{K}_{1221}^{\sigma-\sigma} - \hat{K}_{2112}^{\sigma-\sigma}. \quad (32)$$

One can obtain the "spin flipped" state wave function $|\tilde{\psi}\rangle$

$$\hat{K}''|\psi\rangle = |\tilde{\psi}\rangle. \quad (33)$$

The concurrence C_{II} for the pure state is the mean value of operator \hat{K}''

$$\langle \psi | \hat{K}'' | \psi \rangle = \langle \psi | \tilde{\psi} \rangle = C_{II}. \quad (34)$$

Similarly to the previous case the following relations are valid: $\langle \psi_j | \hat{K}'' | \tilde{\psi}_i \rangle = \delta_{ij}$; $\langle \psi_{i'} | \hat{K}''^2 | \psi_i \rangle = \delta_{ii'}$ and

$$\langle \psi_j | \tilde{\rho} \rho | \psi_i \rangle = \langle \psi_j | \hat{K}'' \rho \hat{K}'' \rho | \psi_i \rangle. \quad (35)$$

The concurrence C [40] expressed through $\tilde{\lambda}_p$ (eigenvalues of matrix $\|\tilde{\rho} \rho\|_{ji}$), arranged in decreasing order, is $C_{II} = \max\{0, \sqrt{\tilde{\lambda}_1} - \sqrt{\tilde{\lambda}_2} - \sqrt{\tilde{\lambda}_3} - \sqrt{\tilde{\lambda}_4}\}$. The expression (35) allows to determine the concurrence C_{II} through the average value of operator \hat{K}''

$$C_{II} = \max\{0, \langle \hat{K}'' \rangle\}. \quad (36)$$

This definition is similar to the obtained expression (29). Consequently, the entanglement [see Ex.(13)] for the pure state is:

$$E(\rho_A) = -\eta_1 \log_2 \eta_1 - \eta_2 \log_2 \eta_2, \quad (37)$$

where η_i are the eigenvalues of matrix $\|\rho_\sigma\|_{ij}$:

$$\rho_\sigma = \begin{pmatrix} \alpha^2 + \beta^2 & \beta\delta + \alpha\gamma \\ \beta\delta + \alpha\gamma & \gamma^2 + \delta^2 \end{pmatrix} \quad (38)$$

with eigenvalues $\eta_{1,2} = \frac{1}{2} \pm \frac{1}{2} \sqrt{1 - C_{II}^2}$. In this case the entanglement is always equal to zero for $C_{II} = 0$. The subsystems with particles with opposite spins are completely disentangled for a pure singlet state in symmetric QDs in the absence of Coulomb interaction.

Let us also introduce the dynamical concurrence $C_I(t)$ and $C_{II}(t)$ for both considered cases in terms of pair correlation functions:

$$C_I(t) = \max\{0, \langle \hat{K}'(t) \rangle\} \quad (39)$$

and

$$C_{II}(t) = \max\{0, \langle \hat{K}''(t) \rangle\}. \quad (40)$$

In this paper we are interested in the initially entangled pure singlet electron state time evolution in the system of coupled QDs with Coulomb correlations, which is the system ground state. Interaction with the reservoir is switched on at the $t = 0$. Our model corresponds to the experimental situation when coupling between vertically aligned strongly interacting QDs and substrate is extremely weak in comparison with the coupling strength with another lead (for example STM tip).

The obtained results for singlet entangled state relaxation are discussed in the next section.

III. RESULTS AND DISCUSSION

The behavior of filling numbers time evolution depends on the initial conditions, which are directly determined by the parameters of the system: energy levels spacing, the Coulomb interaction and interdots coupling values. We also analyzed the concurrence and pair correlation functions time evolution.

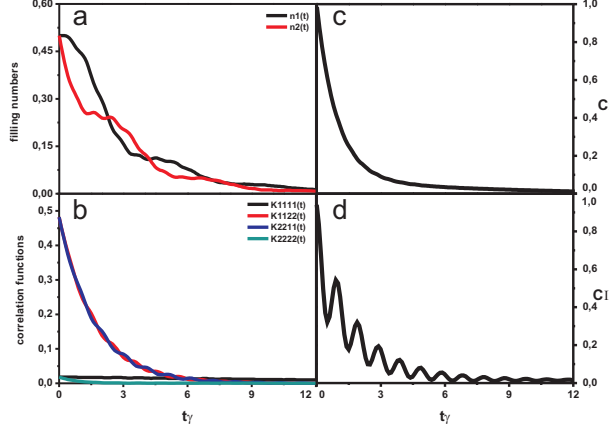


FIG. 2: (Color online) a). Entangled state filling numbers time evolution; b). Entangled state correlation functions time evolution; c). Concurrence C_I time evolution; d). Concurrence C_{II} time evolution. The parameters values: $\varepsilon_1 = \varepsilon_2 = 2.0$; $U_1 = U_2 = 6.0$; $T = 0.6$; $\gamma = 0.3$.

We start by discussing the resonant case: $\varepsilon_1 = \varepsilon_2$ (see Fig.2). At the initial time moment entangled electronic state demonstrates that charge is equally distributed between the energy levels in the QDs [$n_1(0) = n_2(0)$]. Correlation functions $K_{1122}^{\sigma-\sigma}(0)$ and $K_{2211}^{\sigma-\sigma}(0)$ also have the same values which strongly exceed the values of diagonal correlation functions $K_{1111}^{\sigma-\sigma}(0)$ and $K_{2222}^{\sigma-\sigma}(0)$ (see Fig.2b). Such behavior points on the magnetization of the system. For magnetic impurities with definite value of spin projection $S_z = \pm 1/2$, correlators $K_{iii}^{\sigma-\sigma} \ll 1$ (turns to zero). From the other point of view the behavior of pair correlators can be treated as the Hund's rule for the electron filling numbers in coupled QDs with Coulomb correlations. Electron filling numbers $n_1(t)$ and $n_2(t)$ time evolution demonstrate multiple charge redistribution between the QD's energy levels (see Fig.2a).

The dynamical concurrences $C_I(t)$ and $C_{II}(t)$ are demonstrated in the Fig.2c,d. $C_I(t)$ reveals monotonic decreasing during the relaxation processes. Dynamical concurrence $C_{II}(t)$ demonstrates well pronounced oscillations which amplitude decreases with time.

Now we'll discuss the filling numbers time evolution both in the case of the positive ($|\xi| = \varepsilon_1 - \varepsilon_2 > 0$) (see Fig.3) and negative ($|\xi| = \varepsilon_1 - \varepsilon_2 < 0$) (see Fig.4) initial detunings between energy levels in the dots with rather weak Coulomb interaction ($U < |\xi|$).

For the positive detuning at the initial time moment charge in the system is mostly localized on the lower

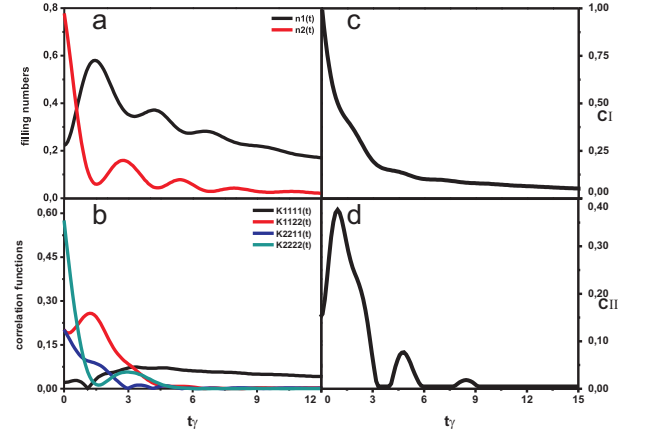


FIG. 3: (Color online) a). Entangled state filling numbers time evolution; b). Entangled state correlation functions time evolution; c). Concurrence C_I time evolution; d). Concurrence C_{II} time evolution. The parameters values: $\varepsilon_1 = 3.5$; $\varepsilon_2 = 2.0$; $U_1 = 1.0$; $U_2 = 1.0$; $T = 0.6$; $\gamma = 0.3$.

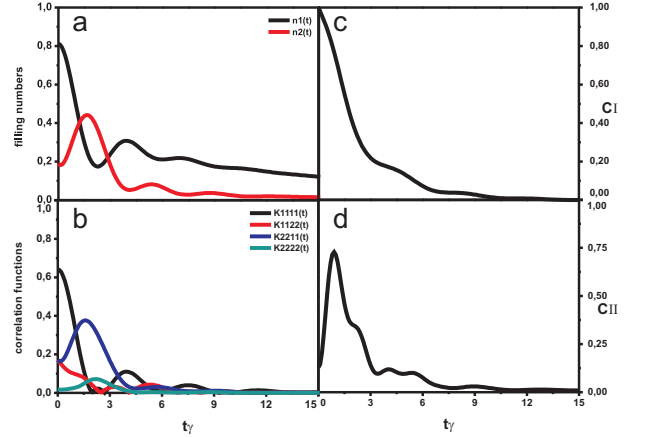


FIG. 4: (Color online) a). Entangled state filling numbers time evolution; b). Entangled state correlation functions time evolution; c). Concurrence C_I time evolution; d). Concurrence C_{II} time evolution. The parameters values: $\varepsilon_1 = 2.0$; $\varepsilon_2 = 3.5$; $U_1 = 0.75$; $U_2 = 0.75$; $T = 0.6$; $\gamma = 0.3$.

energy level [$n_1(0) < n_2(0)$] and the value of correlation function $K_{2222}^{\sigma-\sigma}(0)$ is the largest one in the system (see Fig.3a,b). Filling numbers amplitude $n_1(t)$ continue being smaller than the filling numbers amplitude on the lower energy level $n_2(t)$ during the system evolution until the amplitude of correlation function $K_{2222}^{\sigma-\sigma}(0)$ exceeds amplitudes of other pair correlation functions. During further time evolution the appearance of dynamical inverse occupation occurs and correlation functions $K_{1122}^{\sigma-\sigma}(t)$ and $K_{2211}^{\sigma-\sigma}(t)$ exceed the values of diagonal correlation functions $K_{1111}^{\sigma-\sigma}(t)$ and $K_{2222}^{\sigma-\sigma}(t)$.

In the case of negative detuning at the initial time moment charge in the system is mostly localized on the lower energy level [$n_2(0) < n_1(0)$] and the value of correlation function $K_{1111}^{\sigma-\sigma}(0)$ is the largest one in the system (see Fig.4a,b). Filling numbers amplitude $n_2(t)$ continue be-

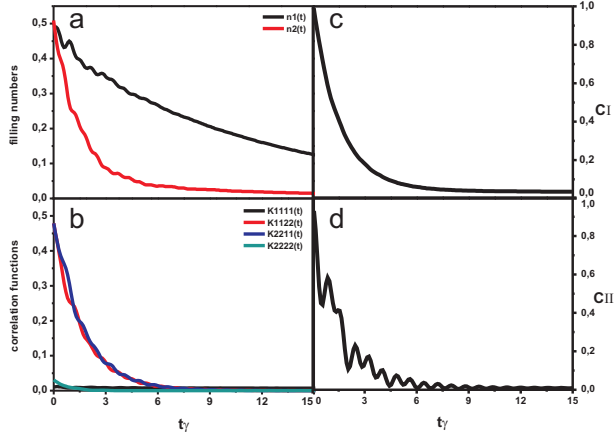


FIG. 5: (Color online) a). Entangled state filling numbers time evolution; b). Entangled state correlation functions time evolution; c). Concurrence C_I time evolution; d). Concurrence C_{II} time evolution. The parameters values: $\varepsilon_1 = 3.5$; $\varepsilon_2 = 2.0$; $U_1 = 6.0$; $U_2 = 6.0$; $T = 0.6$; $\gamma = 0.3$.

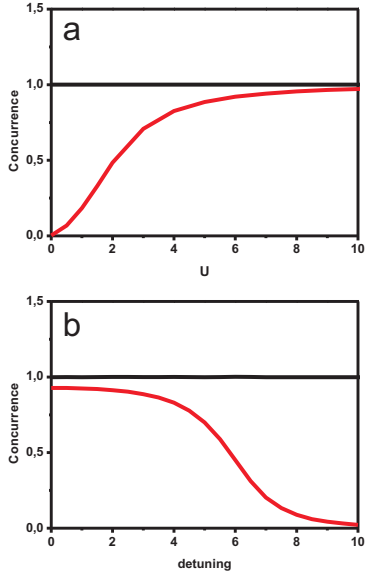


FIG. 6: (Color online) (a) Concurrences $C_I(0)$ (black line) and $C_{II}(0)$ (red line) as a functions of Coulomb interaction value at $t = 0$. Parameters values: $\varepsilon_1 = 3.5$; $\varepsilon_2 = 2.0$; $T = 0.6$; $\gamma = 0.3$. (b) Concurrences $C_I(0)$ (black line) and $C_{II}(0)$ (red line) as a functions of energy levels detuning at $t = 0$. The parameters values: $U_1 = 6.0$; $U_2 = 6.0$; $T = 0.6$; $\gamma = 0.3$.

ing the smaller one during the system evolution until the amplitude of correlation function $K_{1111}^{\sigma-\sigma}(0)$ is the largest one. Relaxation reveals the appearance of dynamical inverse occupation, which takes place when correlation function $K_{2211}^{\sigma-\sigma}(t)$ exceeds the values of other correlation functions. Further time evolution is governed by the correlation function $K_{1111}^{\sigma-\sigma}(t)$ and demonstrates the absence of dynamical inverse occupation.

The dynamical concurrences $C_I(t)$ and $C_{II}(t)$ are

demonstrated in the Fig.3c,d and Fig.4c,d. $C_{II}(t)$ reveal non-monotonic behavior, while $C_I(t)$ monotonically decreases. For both signs of initial detuning concurrence $C_{II}(t)$ rapidly reaches maximum value (formation of a peak) at particular time moment when the electron density is equally distributed between the QDs (see Fig.3a and Fig.4a). For positive initial detuning $C_{II}(t)$ further time evolution reveals non-monotonic behavior and $C_{II}(t)$ even turns to zero at particular time intervals. This means that the system switches between entangled and unentangled states. For negative initial detuning $C_{II}(t)$ also demonstrates non-monotonic behavior, but the system becomes unentangled only at the final stage of charge relaxation.

Localized charge relaxation in the case of QDs with strong Coulomb interaction ($U > |\xi|$) is depicted in the Fig.5. At the initial time moment entangled electronic state demonstrates that charge is quite equally distributed between the energy levels in the QDs [$n_1(0) \leq n_2(0)$]. Correlation functions $K_{1122}^{\sigma-\sigma}(0)$ and $K_{2211}^{\sigma-\sigma}(0)$ have the same values which strongly exceed the values of diagonal correlation functions $K_{1111}^{\sigma-\sigma}(0)$ and $K_{2222}^{\sigma-\sigma}(0)$ (see Fig.5b). Electrons filling numbers $n_1(t)$ and $n_2(t)$ time evolution demonstrates the formation of dynamical inverse occupation during the relaxation process except the initial time moment (see Fig.5a). Similarly to the case when resonant tunneling between the QDs occurs, non-diagonal correlation functions $K_{1122}^{\sigma-\sigma}(t)$ and $K_{2211}^{\sigma-\sigma}(t)$ dominate in the charge relaxation process (see Fig.5a). As we mentioned above this effect can be treated as the fulfillment of the Hund's rule in coupled QDs with Coulomb correlations.

The dynamical concurrence $C_I(t)$ amplitude decreases monotonically with the decreasing of localized charge amplitude in the system. Time evolution of $C_{II}(t)$ demonstrates well pronounced oscillations which amplitude decreases with time.

Let us now discuss the behavior of concurrence at the initial time moment. For initial ground two-electron state in coupled QDs $C_I(0)$ and $C_{II}(0)$. Concurrences $C_I(0)$ (black line) and $C_{II}(0)$ (red line) as a functions of Coulomb interaction value and detuning between energy levels in the QDs are depicted in the Fig.6a and Fig.6b correspondingly. Concurrence $C_I(0)$ is always equal to unity for all values of initial detuning and Coulomb interaction (see Fig.6a and Fig.6b black line). $C_{II}(0)$ (red line) in the absence of Coulomb interaction is equal to zero, the system is in the pure unentangled state. The increasing of Coulomb interaction value results in the growth of the $C_{II}(0)$ ($C_{II}(0) \rightarrow 1$ for $U \rightarrow \infty$) (see Fig.6a red line). Consequently, Coulomb correlations in the system for infinitely large U lead to the formation of the fully entangled Bell's electronic state at the initial time moment for coupled QDs. Fig.6b (red line) demonstrates that the concurrence $C_{II}(0)$ monotonically decreases with the detuning growth. The entanglement disappears when energy levels spacing strongly exceeds the Coulomb interaction value.

IV. CONCLUSION

We demonstrated that for arbitrary mixed state the concurrence can be determined from the average value of particular combinations of localized electrons pair correlation functions. We obtained the closed system of equations for time evolution of the localized electrons filling numbers and pair correlation functions which exactly takes into account all order correlations for localized electrons.

From time dependence of electrons filling numbers and pair correlation functions one can follow the time evolution of concurrence and entanglement during the relaxation process. We analyzed different possible ways to divide the investigated system into two entangled subsystems. We have found special regimes when dynamical concurrence demonstrates non-monotonic behavior during the time evolution.

We revealed the appearance of dynamical inverse occupation of the QDs energy levels and demonstrated that for large values of Coulomb interaction non-diagonal pair correlation functions always exceed the diagonal ones.

When on-site Coulomb repulsion is smaller than the energy levels detuning, correlation function of two electrons with opposite spins localized in the QD with the lower energy level exceeds all other correlation functions until the dynamical inverse occupation occurs due to the relaxation process.

We also analyzed the dependence of initial value of concurrence on the system parameters: Coulomb correlations value and energy levels detuning. We revealed that concurrence $C_{II}(0)$ for large Coulomb interaction values ($U \rightarrow \infty$) is close to unit for the finite value of detuning. Concurrence $C_I(0)$ is always equal to unity for all values of initial detuning and Coulomb interaction. We also demonstrated the validity of Hund's rule for the two coupled single level QDs when Coulomb interaction value is larger than the energy levels detuning. Our results open up further perspectives in solid state quantum information based on the controllable dynamics of the entangled electron states.

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